Positron irradiation effect on positronium formation in gamma-irradiated LDPE and unplasticized PVC

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Abstract

Positron irradiation effects on positronium formation in low-density polyethylene (LDPE), gamma-irradiated LDPE and unplasticized PVC (UPVC) are studied. At least in one of the three different measurements, i.e., prolonged positron annihilation measurement at room temperature, low temperature in darkness and subsequent measurement under light, changes in *o*-Ps intensity are observed in non-irradiated LDPE and gamma-irradiated LDPE. While in UPVC, change in *o*-Ps intensity is hardly observable in all the above-mentioned three measurements. Reduction of *o*-Ps intensity by light indicates that positronium formation via the recombination of a positron and a trapped electron exists in LDPE and gamma-irradiated LDPE. The absence of light bleaching effect, together with the fact that the value of *o*-Ps intensity in heating and cooling process of a thermal circle is nearly the same, indicates that in UPVC, positronium can not be formed through trapped electron mechanism. This study highlights the speciality of positronium formation in UPVC, positronium is formed exclusively by the recombination of electron-positron pairs with short separations.

Keywords: Positron irradiation effect; Positronium formation; Temperature; Light

1 Introduction

Positron annihilation technique has turned out to be a unique tool to detect subatomic free-volume in polymeric materials (Jean et al., 2003). In polymer, positronium, the bound state of a positron and an electron, can be formed and annihilates in free volume. Positronium can be divided into two categries: singlet para-positronium (*p*-Ps) with the spins of the positron and electron antiparallel and triplet ortho-positronium (*o*-Ps) with parallel spins. In polymer, *p*-Ps undergoes self-annihilation (positron in *p*-Ps annihilates with its own partner electron) with a lifetime of 125 ps, just like in vacuum. However, during its lifetime in a free volume *o*-Ps will intereact with the molecules of the surrounding medium, pick up an electron and annihilate with it other than its own partner, so called "pick-off annihilation". Therefore, the annihilation parameters of *o*-Ps are directly related to the characteristics of free volume. For a long time, it was commonly accepted that *o*-Ps lifetime depended on the size of free volume (Tao, 1972), and *o*-Ps intensity was the direct measure of the number density of free volumes (Nakanishi et al., 1988). However, Kindl (Kindl and Reiter, 1987) and Welander (Welander and Maurer, 1992) reported the gradual increase and gradual decrease of *o*-Ps intensity with the measurement time in polymers at low temperature and at room temperature, respectively. Until then, researchers realized that free volume was not the only factor that affected positronium formation in polymers. Positron irradiation effect, the irradiation damage induced by positrons emitted from ^{22}Na source, should be paid special attention in polymer studies.

Many researchers have studied positron irradiation effect in various polymers, it is found that such effects depend on temperature, polymer type, and polymer state. Now it is well accepted that at room temperature, the decrease of *o*-Ps intensity in PE, PP and other polymers is due to free radicals introduced by positron irradiation. Free radicals are easily induced by high-energy radiation such as gamma rays and electron beams. Positrons emitted from the ^{22}Na positron source with the average energy of approximately 220 keV, can easily induce free radicals in polymers. Free radicals have a strong affinity to electrons and act

as a scavenger of electrons. According to the spur model, free radicals can suppress positronium formation (Chen et al., 2001). Ps formation probability depends on the number of available electrons around the end of positron spur. Free radicals accumulate with elapsed time, thus *o*-Ps intensity decreases. While the enhancement of positronium formation at low temperature is related to a new positronium formation mechanism proposed by Hirade (Hirade et al., 2000). According to the Ps formation model, at very low temperature (≪Tg), shallow potential is created at a site surrounded by mobility-frozen molecules. Some of the excess electrons produced by positron irradiation are trapped in these shallow potentials. If a free positron finds one of the trapped electrons before its annihilation, the positron will pick up a trapped electron to form Ps. The enhancement of Ps formation is due to the accumulation of trapped electrons with elapsed time. Moreover, the binding energy of the electrons on the trapping sites is 0.5–3 eV (Hirade et al., 2000). Therefore, the visible light, with energy band from 1.6 to 3.2 eV, can quench the trapped electrons. Hence the light bleaching effect on Ps formation should be a good evidence for the existence of the special Ps formation mechanism at low temperature (Hirade et al., 2007).

Besides temperature, positron irradiation effect is also dependent on polymer type and polymer state. It is found that in non-polar polymers like PE, PP, PS (Peng et al., 1996; Suzuki et al., 1995, 1996; Uedono et al., 1997), the decrease or increase of positronium yield with elapsed time is striking, while in polar polymers, like CPE (chlorinated polyethylene) and ~~PMMA(~~PMMA (Qi et al., 2000; Wang et al., 2001), positron irradiation effect becomes non-obvious. Different behaviours of non-polar and polar polymers are related to different positronium formation process. In non-polar polymers like LDPE, free positrons have large mobility and positronium can be formed via recombination of electron-positron pairs with both large and short initial separations (Wang et al., 2001; Yu et al., 2006). Among them, the former is easily affected by factors such as electric ~~fieldpolar~~ field, polar groups and free radicals. However, the latter is hardly affected by above factors. While in polar polymers like CPE, positronium is formed exclusively by the recombination of electron-positron pairs with short separations.

UPVC is a polar polymer which contains C-Cl groups, *o*-Ps intensity in it is very low (less than 5%). It will be very interesting to see whether positron irradiation effect can affect positronium formation in UPVC. In addition, after non-polar polymers are irradiated by gamma-rays, polar groups and free radicals are induced, then the behaviour of positronium yield in irradiated polymers becomes complicated. For example, at low temperature, in heavily irradiated PE, *o*-Ps intensity has no significant change with time, some researchers (Yu et al., 2005) said that positronium formation of a positron and a trapped electron is strongly suppressed by the scavenge of excess electrons by free radicals created by gamma irradiation, while others (Suzuki et al., 2000) insist that the pre-irradiation just slows down the process of positronium formation. Focusing on the above problems, positron irradiation effect in heavily gamma-irradiated LDPE is studied. For comparison, the result of non-irradiated LDPE is also included. Since the light bleaching effect is the unique characteristic of positronium formation of a positron and a trapped electron, it is used to verify whether the special Ps formation mechanism exists or not.

In this paper, at both room temperature and low temperature, positron irradiation effects on positronium formation in LDPE, gamma-irradiated LDPE, and polar polymer UPVC are studied. At low temperature, light bleaching effect is used to verify if the special positronium formation mechanism exists in these polymers.

2 Experiment

Polymer samples used in this study were purchased from Goodfellow (Cambridge, UK), including low-density polyethylene (LDPE) and unplasticized Polyvinyl chloride (UPVC). Polymer films, cut into plates of 10×10 mm^2 and put into glass vessels, were irradiated by a ^{60}Co Gamma cell in China Institute of Atomic Energy. LDPE samples were irradiated with absorbed dose of 100, 200, 1000, 2000 kGy. For convenience, the sample is referred by sample name and its irradiation dose, for example, LDPE films irradiated by gamma rays in air with a total dose of 1000 kGy is denoted by LDPE1000.

Positron annihilation lifetime measurements (PALS) were performed with a fast-slow coincidence system using the BaF_2 scintillators. PALS system had a resolution of 196 ps (full width at half maximum for the ^{60}Co prompt γ-rays, under ^{22}Na window settings). Low temperature measurements were carried out using an 8 K closed cycle Helium fridge. One hour was required to attain the lowest temperature of about 10 K. A temperature controller (Lakeshore 330) was used to maintain the sample temperature at the set value. The sample chamber was specially designed with an optical window, by which the light from laser torch can arrive at the position where samples located. To study positron irradiation effect at room temperature, the samples were continuously measured for about 90 h (the spectrum was saved every two hours, resulting in 1.6 million events). To study positron irradiation effect at low temperature, Ssamples were first kept at 10 K in darkness, PALS was continuously measured. Besides PALS, Coincidence Doppler Broadening (CDB) measurements were also carried out to monitor the chemical environment around positron annihilation sites, the details of CDB measurements can be found elsewhere (Rahman et al., 2014). In the first 20 h, PALS and CDB measurements were measured simultaneously, but it was found that the statistics of obtained CDB spectrum was too low. So after 52 h, when *o*-Ps intensity got to its saturation value, PALS measurement stopped, and the time from 52 to 68 h was used for CDBS measurement (curve A2). From 72 h to 100h, green light was applied, PALS was continuously measured for 12 h. At 84 h, when *o*-Ps intensity again got to its saturation value, PALS measurement was replaced by CDB measurement (curve B1). In these two stages, after the CDB measurement finished, two more PALS spectra were collected to monitor the change of *o*-Ps intensity. From 100 h, green light was replaced by white light from flashlight. 6 h later, the flashlight was turned off. The Helium fridge was also turned off, and the sample gradually recovered to room temperature. Low-temperature measurement scheme is shown in Fig. 1.

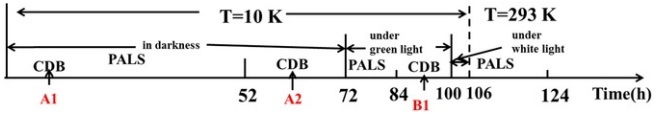

Fig. 1 The scheme of low temperature measurement.

alt-text: ~~Fig.1~~Fig. 1

3 Results and discussion

3.1 ~~positron~~Positron irradiation effect on positronium formation at room temperature

The variations of *o*-Ps lifetime and intensity with the extension of measurement time at room temperature for non-irradiated LDPE, 1000 kGyγ-irradiated LDPE and non-irradiated UPVC are shown in Figs. 2–4. In all the three samples, *o*-Ps lifetime τ_3 keeps nearly constant value in the whole measurement, 2.5 ns and 2.38 ns for non-irradiated and 1000 kGyγ-irradiated LDPE, 1.55 ns for UPVC. In comparison with non-irradiated LDPE, The reduction of τ_3 in LDPE1000 indicates the shrinkage of free volume mainly due to crosslinking of polymer chains induced by gamma irradiation (Al-Qaradawi et al., 2003). It is clear that *o*-Ps lifetime τ_3 is influenced by gamma irradiation but not by positron irradiation. In contrast, *o*-Ps intensity can be inhibited by both gamma irradiation and positron irradiation. After 98 h′ measurement, the *o*-Ps intensity in non-irradiated LDPE is lowered from the initial value of 24.5% to about 23%, with a decreasement of 6%. The decreasement of *o*-Ps intensity is fast in the first 30 h, thereafter the decreasement becomes very slow. It seems that the *o*-Ps intensity has reached its saturation value. Interestingly, *o*-Ps intensity in LDPE1000 and UPVC shows similar behaviour. There are no observable changes in *o*-Ps intensity, which keeps around 15.6% in LDPE1000 and 4.3% in UPVC.

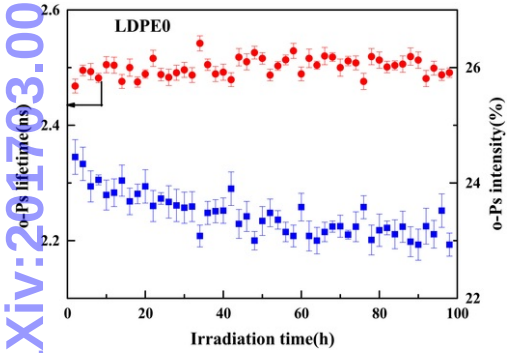

Fig. 2 The variation of *o*-Ps lifetime and intensity with time at RT in non-irradiated LDPE. In the figure, red solid circles and blue solid squares represent *o*-Ps lifetime and intensity, respectively. Symbols in Figs. 3, 4, 6, 8, 9 and 10 are the same as in Fig. 2.

alt-text: ~~Fig.2~~Fig. 2

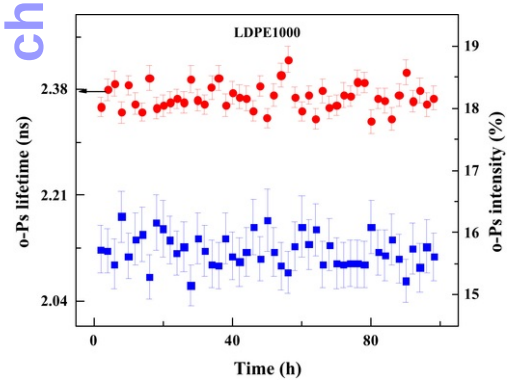

Fig.3 The variation of *o*-Ps lifetime and intensity with time at RT in gamma-irradiated LDPE.

alt-text: Fig.3

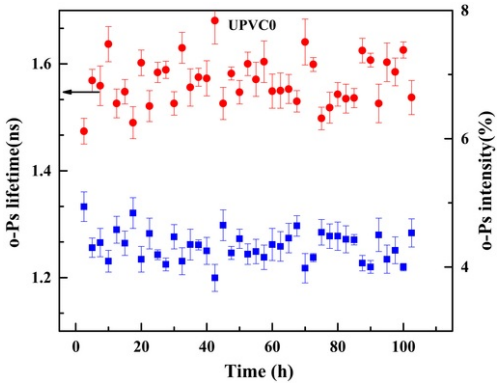

Fig. 4 The variation of *o*-Ps lifetime and intensity with time at RT in UPVC.

alt-text: ~~Fig.4~~Fig. 4

In polymers, positronium can be formed via recombination of positron with one of the excess electrons. Free radicals induced by positron irradiation and gamma irradiation can scavenge electrons created by positron and thus suppress positronium formation. In non-irradiated LDPE, positronium can be formed via recombination of electron-positron pairs with both large and short initial separations. Since positronium formed via electron-positron pairs with large initial separation is easily affected by factors such as electric field, polar groups and free radicals. As the result of suppression effect by free radicals, the decrease of *o*-Ps intensity with elapsed time in non-irradiated LDPE is mainly resulted from positronium formed via electron-positron pairs with large initial separations.

UPVC is a polar polymer, containing C-Cl groups, which is associated with a special positron capture mechanism: electrons produced by positron irradiation undergo dissociative attachment to C-Cl groups and resultant chloride ions capture positrons. In heavily gamma irradiated LDPE, FTIR results in Fig. 5 show that carbonyl groups appear. Oxygen atoms on carbonyl groups can also capture positrons. As a result, both UPVC and heavily gamma irradiated LDPE contain polar groups. Polar groups dramatically decrease the mobility of free positrons (Zheng et al., 1998), thus positronium may be formed exclusively by the recombination of electron-positron pairs with short separations. On the other hand, since the reduction of *o*-Ps intensity is the result of suppression effect from free radicals, free radicals in these two polymers may not work effectively at room temperature. According to the study by Qi (Qi et al., 2015), free radicals in PMMA are not stable at RT, which results in the absence of decrease in *o*-Ps intensity with elapsed time. However, when the temperature falls to 225 K, there is a significant decline in *o*-Ps intensity with time. Here, at room temperature, *o*-Ps intensity in LDPE1000 and UPVC show similar behaviour to PMMA, i.e. no observable changes with time. It is inferred that free radicals in LDPE1000 and UPVC may show similar behaviour to PMMA. Further analysis will be conducted in combination with results at low temperature.

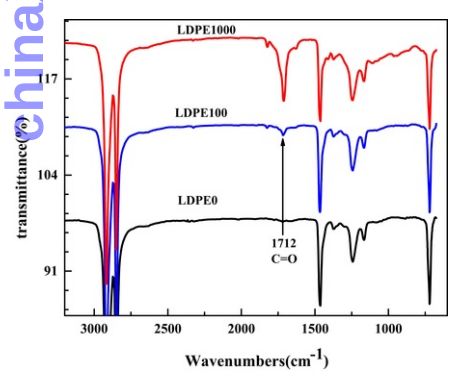

Fig. 5 FTIR spectra of gamma irradiated LDPE samples.

alt-text: ~~Fig.5~~Fig. 5

3.2 ~~positron~~Positron irradiation effect on positronium formation at low temperature

At low temperature (T=10 K), the behaviour of *o*-Ps intensity with time is very different from that at RT. As shown in Fig. 6, the *o*-Ps intensity of non-irradiated LDPE increases from 18% to about 30%, with an increasement of 67% during 50 h′ irradiation in darkness. *o*-Ps intensity increases rapidly within initial 40 h and later tends to saturate. From 52 h to 68h, PALS measurement paused, instead, CDB measurement was carried out. After 16 h′ CDB measurement, it is found that *o*-Ps intensity in LDPE is still around 30%, confirming *o*-Ps intensity has reached its saturation value. Next, when green light is applied, *o*-Ps intensity sharply decreases down to 19% in two hours. After green light is applied for 4 h, *o*-Ps intensity reduces to 16% and thereafter remains nearly unchanged. After 12 h′ measurement under green light, PALS measurement paused, instead, CDB measurement was carried out. After the CDB measurement, it is found that *o*-Ps intensity in LDPE is still around 16%, confirming *o*-Ps intensity has reached its saturation value. Note that the saturation value of *o*-Ps intensity under light is lower than the initial value in darkness at 10 K, indicating that positronium formed via the recombination of a positron and a trapped electron appears in the first two hours, or in other words, the initial *o*-Ps intensity in dark should be 16% instead of 18%. After 25 h's measurement under green light, white light is applied to check if all the trapped electrons are smeared out. It is seen that there is little change in *o*-Ps intensity for LDPE under white light for 6 h, proving that trapped electrons have already been completely eliminated.

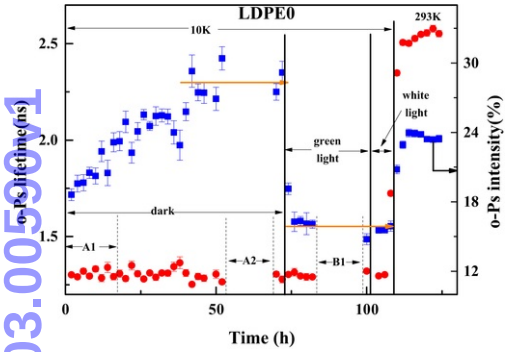

Fig. 6 The variation of *o*-Ps lifetime and intensity with time and the influence of light at 10 K in non-irradiated LDPE.The lines in orange are just guide for eyes.

alt-text: ~~Fig.6~~Fig. 6

During the measurement, three CDB spectra are collected: the one collected in the first 20 h, region A1 in Fig. 6; the one collected in the area where *o*-Ps intensity is saturated, Region A2; the one collected in the area where all the trapped electrons are eliminated, region B1. Using the CDB spectrum measured at RT as the reference sample, the ratio curves of CDB spectrum in region A1 and A2 demonstrate the temperature effect, and the ratio curve of CDB spectrum in region B1 demonstrates the light bleaching effect, as shown in Fig. 7. It can be seen that all the three curves show a peak between 5 and 15 (10^{-3} m_0c), the specific peak from positron annihilation with the core electrons of carbon atom (Suzuki et al., 2003). In region A1, positron annihilation at LT is not so different from that at RT, (*o*-Ps intensity is 24.5% at RT and 18% at LT) thus the peak intensity is low. In region A2, even though there is a large enhancement in *o*-Ps intensity, the peak intensity is still very low, indicating that there are few interactions between positrons and the core electrons of carbon atoms, and many Ps disappear through pick-off annihilation with valance electrons with a low momentum. On the contrary, the peak intensity of CDB ratio curve in region B1 is very high. This is because of the large reduction of Ps formation, thus a large fraction of free positron annihilation with core electrons of carbon atoms. (For gamma irradiated LDPE and UPVC, CDB spectra are also collected, however, CDB results show no difference, therefore these results are not given).

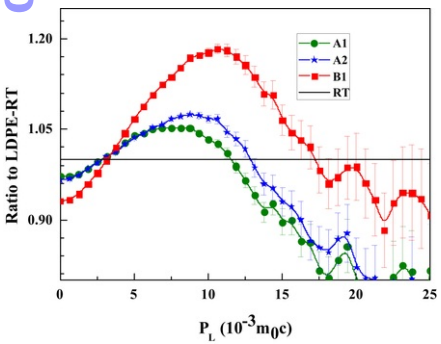

Fig. 7 CDB ratio curve of non-irradiated LDPE. Curves A1 and A2 show temperature effect, curve B1 shows light-bleaching effect. Curve RT is obtained at room temperature (T=293 K), while curves A1, A2 and B1 are the results obtained at low temperature

(T=10 K). Among them, A1 represents CDB spectrum measured in the first 20 h in darkness, A2 and B1 represent CDB spectra measured in the stage where *o*-Ps intensity gets to its saturation value in darkness and under green light, respectively.

alt-text: ~~Fig.7~~Fig. 7

Fig. 8 shows positron irradiation effects at 10 K in gamma-irradiated LDPE samples, LDPE200 and LDPE2000. In both samples, after 50 h′ measurement in darkness, *o*-Ps intensity shows no obvious changes. However, when green light is applied, *o*-Ps intensity decreases. For LDPE200, *o*-Ps intensity decreases from the value in darkness 15.5-to 14%. For LDPE2000, *o*-Ps intensity decreases from the value in darkness 14.3-to 13%. Since light bleaching effect is the unique characteristic of positronium formed by a positron and a trapped electron, the reduction of *o*-Ps intensity by light indicates the existence of the special positronium formation mechanism at low temperature in gamma-irradiated LDPE. Radicals and oxidized species induced by gamma irradiation just slow down the process of positronium formation of a positron and a trapped electron, *o*-Ps intensity still can get obvious increasement with prolonged measurement time.

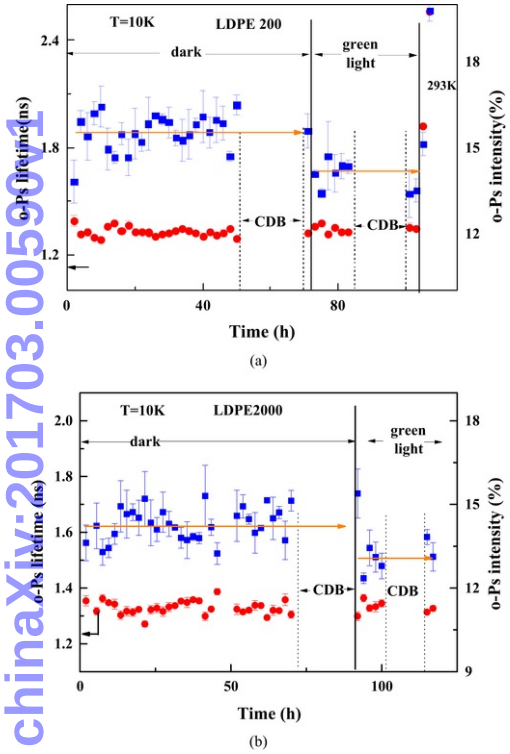

Fig. 8 The variation of *o*-Ps lifetime and intensity with time and the influence of light at 10 K in gamma-irradiated LDPE samples: (a) LDPE200 (b) LDPE2000. The lines in orange are just guide for eyes.

alt-text: ~~Fig.8~~Fig. 8

Fig. 9 shows positron irradiation effect at 10 K in UPVC. In darkness, UPVC shows similar behaviour to gamma-irradiated LDPE, *o*-Ps intensity keeps nearly constant value during 50 h′ measurement. Interestingly, *o*-Ps intensity in UPVC still stays unchanged when green light is applied. The absence of both increasement in darkness and decreasement under green light for *o*-Ps intensity in UPVC suggests that positronium formed via the recombination of a positron and a trapped electron may not exist.

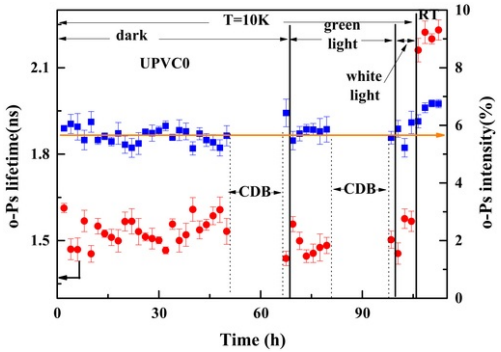

Fig. 9 The variation of *o*-Ps lifetime and intensity with time and the influence of light at 10 K in UPVC. The lines in orange are just guide for eyes.

alt-text: ~~Fig.9~~Fig. 9

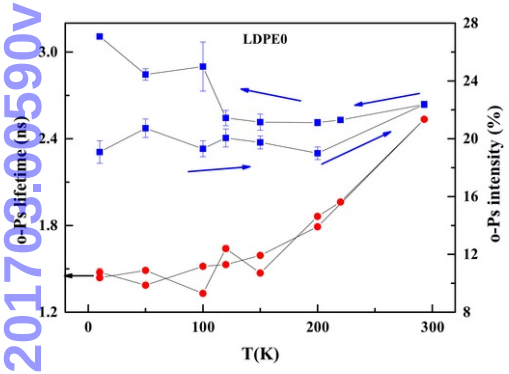

Fig. 10 The variation of *o*-Ps lifetime and intensity with temperature in a thermal circle 10 K-293 K-10 K in LDPE.

alt-text: ~~Fig.10~~Fig. 10

To further confirm the speciality of positronium formation in UPVC, the variations of *o*-Ps lifetime and intensity with temperature in UPVC in a thermal circle 50 K-293 K-50 K in darkness are given in Fig. 11, in comparison with the result of LDPE (10 K-293 K-10 K) in Fig. 10. The changes of *o*-Ps lifetime mainly reflect the expansion and the shrinking of free volume. The thermal circle is accompanied by the rearrangement of polymer chains. Since the rearrangement of polymer chains in the process of arising and lowing temperature is reversible, the dimension of free volume, thus *o*-Ps lifetime in heating process is roughly the same with that in cooling process. The behaviour of *o*-Ps lifetime is the similar in LDPE and UPVC. Below we will only focus on the variation of *o*-Ps intensity. For non-polar polymer LDPE, *o*-Ps intensity increases slowly during the elevation of temperature from 10 K to 150 K, it decreases to some extent at 200 K and reaches to a relatively high value at room temperature. In cooling process, *o*-Ps intensity decreases from RT to 200 K, thereafter, it begins to increase significantly, finally gets the value of 30% at 10 K. Interestingly, the value of *o*-Ps intensity after a thermal circle is nearly the same as that after 52 h′ measurement in darkness at 10 K (Fig. 6). It takes about 33 h to finish the thermal circle, temperature effect is accompanied with time effect. Shallow trapped electrons accumulate with time, thus *o*-Ps intensities between 150 K and 10 K in cooling process are much higher than that in heating process. While in UPVC, the value of *o*-Ps intensity in cooling process and in heating process is nearly the same in the whole temperature range. This may serve as another proof that the special positronium formation at low temperature is not exist in UPVC.

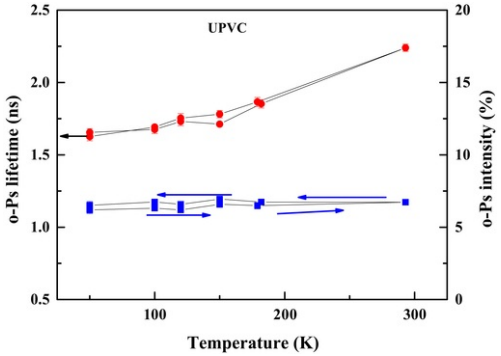

Fig. 11 The variation of *o*-Ps lifetime and intensity with temperature in a thermal circle 50 K-293 K-50 K in UPVC.

alt-text: ~~Fig.11~~Fig. 11

It is worth noting that at low temperature, positronium formation through the mechanism proposed by Hirade mainly occurs to the electron-positron pairs with large separations. In UPVC, the absence of positronium formation of a positron and a trapped electron, verifies that positronium is formed exclusively by the recombination of electron-positron pairs with short separations. While in gamma irradiated LDPE, the appearance of positronium formation of a positron and a trapped electron, indicates that positronium can also be formed by the recombination of electron-positron pairs with large separations. Thus, the absence of decrease of *o*-Ps intensity at room temperature should ascribe to free radicals. It is likely that in gamma irradiated LDPE, free radicals are not stable at RT, just like in PMMA.To further verify this conclusion, positron irradiation effect at near room temperature(T<293 K) in gamma irradiated LDPE can be done.

4 Conclusion

The effects of positron irradiation on positronium formation in LDPE, gamma-irradiated LDPE and UPVC, both at room temperature and at low temperature are studied. In LDPE, *o*-Ps intensity decreases with time at room temperature and increases with time in darkness at low temperature, light can result in sharply decrease of *o*-Ps intensity. In addition, in a thermal circle 10 K-293 K-10 K, *o*-Ps intensity increases significantly in cooling process below 150 K. While in heavily gamma-irradiated LDPE and UPVC, neither the decrease of *o*-Ps intensity with time at room temperature nor the increase of *o*-Ps intensity with time in darkness at low temperature appears. *o*-Ps intensity shows no observable changes with elapsed time. However, the effect of light on positronium formation is different in heavily gamma-irradiated LDPE and UPVC. In the former, application of light leads to the decrease of *o*-Ps intensity, in contrast to the absence of changes in *o*-Ps intensity in the latter. The appearance of light bleaching effect suggests that the special positronium formation mechanism at low temperature exist in heavily gamma-irradiated LDPE. The absence of light bleaching effect, together with the fact that the value of *o*-Ps intensity in cooling process and in heating process is nearly the same in the whole temperature range indicates that in UPVC, positronium can not be formed through the combination of a positron and a shallow trapped electron. This study highlights the speciality of positronium formation in UPVC, positronium is formed exclusively by the recombination of electron-positron pairs with short separations. In addition, at RT, the absence of positron irradiation effect on positronium formation in UPVC and in gamma irradiated LDPE is due to different reasons. The former is attributed to positronium itself, which is hardly influenced by free radicals and temperature, the latter is due to free radicals. It is likely that in gamma irradiated LDPE, free radicals are not stable at RT, just like in PMMA.

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References

Al-Qaradawi I.Y., Britton D.T., Abdel-Hady E.E., Abdulmalik D.A., Al-Shobaki M.A. and Minani E., Positron annihilation studies of the effect of gamma irradiation dose in polymers, ~~*Radiat Phys Chem*~~*Radiat. Phys. Chem.* **68**, 2003, 457–461.

Chen Z.Q., Suzuki T., Kondo K., Uedono A. and Ito Y., ~~Free Volume~~free volume in ~~Polycarbonate Studied~~polycarbonate studied by Positron ~~Annihilation: Effects~~annihilation: effects of ~~Free Radicals~~free radicals and ~~Trapped Electrons~~trapped electrons on Positronium ~~Formation. Jpn~~formation, *Jpn. J. Appl. Phys.* **40**, 2001, 5036–5040.

Hirade T., Maurer F.H. and Eldrup M., Positronium formation at low temperatures: the role of trapped electrons, *Radiat. Phys. Chem.* **58**, 2000, 465–471.

Hirade T., Suzuki N., Saito F. and Hyodo T., Reactions of free positrons in low temperature polyethylene: delayed Ps formation and positron localization, ~~*Physica. Status. Solidi. (c)*~~*Physica Status Solidi (C)* **4**, 2007, 3714–3717.

Jean Y., Mallon P. and Schrader D.M., Principles and ~~applications~~Applications of ~~positron~~Positron & ~~positronium chemistry~~Positronium Chemistry, 2003, World Scientific Publishing Company Incorporated.

Kindl P. and Reiter Q., Investigations on the ~~Low-Temperature Transitions~~low-temperature transitions and ~~Time-Effects~~time effects of ~~Branched-Polyethylene~~branched polyethylene by the Positron ~~Lifetime-Technique~~lifetime technique, ~~*Physica. Status. Solidi.*~~*Physica Status Solidi (a)* **104**, 1987, 707–713.

Nakanishi H., Wang S., Jean Y. and Sharma S., Positron annihilation studies of fluids, 1988, World Science; Singapore, 292.

Peng Z., Itoh Y., Li S. and Wang S., Positron irradiation effects during positron annihilation lifetime measurement in polymers, ~~*Physica. Status. Solidi.*~~*Physica Status Solidi (a)* **155**, 1996, 299–303.

Qi C., Wei W., Wu Y., Zhang S., Haijun W., Li H., Wang T. and Yan F., Effect of polymer polarity on the positronium formation, *J. Polym. Sci. B* **38**, 2000, 435–442.

Qi N., Chen Z.Q. and Uedono A., Molecular motion and relaxation below glass transition temperature in poly (methyl methacrylate) studied by positron annihilation, *Radiat. Phys. Chem.* **108**, 2015, 81–86.

Rahman A.Z.M.S., Li Z., Cao X., Wang B., Wei L., Xu Q. and Atobe K., Positron annihilation study of vacancy-type defects in fast-neutron-irradiated ~~MgO·nAl2O3~~$MgO{\cdot}nAl_2O_3$, *Nucl. Instr. Meth. B* **335**, 2014, 70–73.

Suzuki T., He C., Shantarovich V., Kondo K., Hamada E., Matso M., Ma L. and Ito Y., The influence of radiation and light on Ps formation in PMMA and PE studied by coincidence Doppler-broadening spectroscopy, *Radiat. Phys. Chem.* **66**, 2003, 161–166.

Suzuki T., Ito Y., Kondo K., Hamada E. and Ito Y., Radiation effect on positronium formation in low-temperature polyethelene, *Radiat. Phys. Chem.* **58**, 2000, 4.

Suzuki T., Miura T., Oki Y., Numajiri M., Kondo K. and Ito K., Positron irradiation effects on polypropylene and polyethylene studied by positron annihilation, *Radiat. Phys. Chem.* **45**, 1995, 7.

Suzuki T., Oki Y., Numajiri M., Miura T., Kondo K., Oshima N. and Ito Y., Radiation effect on polypropylene studied by the relaxational behaviour at low temperature using positron annihilation, *Polymer* **37**, 1996, 5521–5524.

Tao S., Positronium annihilation in molecular substances, ~~*J. Chem.*~~ *J. Chem. Phys.* **56**, 1972, 5499–5510.

Uedono A., Kawano T., Tanigawa S., Ban M., Kyoto M. and Uozumi T., Transition and relaxation processes of polyethylene, polypropylene, and polystyrene studied by positron annihilation, *J. Polym. Sci. B* **35**, 1997, 1601–1609.

Wang C., Kobayashi Y., Zheng W., Zhang C., Nagai Y. and Hasegawa M., Positronium formation in a polymer blend of polyethylene and chlorinated polyethylene, *Phys. Rev. B* **63**, 2001.

Welander M. and Maurer F.H., Decrease in o-Ps formation in polymers during positron lifetime measurements, *Mater. Sci. Forum* 1992, 1811–1814.

Yu R.S., Ito Y., Suzuki T., Shantarovich V. and Kondo K., External electric field effect on positronium formation in some polar and nonpolar polymers at different temperatures, *Chem. Phys.* **327**, 2006, 278–282.

Yu R.S., Suzuki T., Djourelov N., Kondo K., Ito Y. and Shantarovich V., Positron annihilation lifetime and coincidence Doppler broadening study of γ-irradiated polyethylene, *Chem. Phys.* **313**, 2005, 63–69.

Zheng W., Kobayashi Y., Hirata K. and Suzuki T., Mobility of positrons in gamma-irradiated polypropylene and polyethylene, *Radiat. Phys. Chem.* **51**, 1998, 269–272.

Highlights

- Positron irradiation effects at RT and 10 K, in LDPE and in UPVC are studied.
- Different factors lead to absence of changes in I_3 at RT in irradiated LDPE and UPVC.
- Light bleaching effect appears in gamma irradiated LDPE.

- Absence of light-bleaching effect in UPVC shows the speciality of Ps formation.
- Positronium formation of a positron and a trapped electron doesn’t exist in UPVC.

Queries and Answers

Query:

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Answer: Yes